



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Takao OHNO, et al.

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For: POLYMETAPHENYLENE ISOPHTHALAMIDE-BASED POLYMER POROUS
FILM, PROCESS FOR ITS PRODUCTION AND BATTERY
SEPARATOR

DECLARATION UNDER 37 C.F.R. 1.132

Commissioner for Patents

Alexandria, VA 22313-1450

Sir:

I, Satoshi NISHIKAWA, c/o TEIJIN LIMITED, Iwakuni
Research Centre, 2-1, Hinodecho, Iwakuni-shi, Yamaguchi
740-0014, Japan do hereby declare:

That I am by profession a chemical engineer
having earned a master's degree in electrochemistry from
Tokyo Institute of Technology in 1998;

That I joined TEIJIN LIMITED in 1998 and from 1998 to the present have engaged in research and development work at the Iwakuni Research Center of TEIJIN LIMITED, I am not an inventor of the invention in the above-identified U.S. application (hereinafter referred to as "present invention" for brevity) but was engaged in the development of lithium ion battery separators from 1998 to the present, and due in part to this background, I have the technical knowledge possessed by persons skilled in the art in the technical field and sufficiently understand the technical content of the present invention;

That, from 1998 to 2006, I was affiliated with the same research section as Mr. Takahiro Daido, a co-inventor the invention disclosed in U.S. Patent No. 6,291,106 (hereinafter referred to as "Daido" for brevity) cited against the claims of the above-identified U.S. application (hereinafter referred to as "present application" for brevity), due in part to this, I also sufficiently understand the technical content of Daido, and I have made many inventions jointly with Mr. Takahiro Daido et al on technology relating to lithium ion batteries such as in U.S. Patent No. 6,818,352, U.S. Patent No. 6,881,438 and U.S. 2005/0,277,026;

That I conducted the following experimental working, and will clarify based on the results that the present invention and the art of Daido differ.

Experimental Working

Experiment Regarding the Present Invention

I prepared a poly-meta-phenyleneisophthalamide (m-aramid)-based polymer porous film according to the present invention.

Specifically, I prepared a dope comprised of a dimethyl acetoamide (DMAc) solution containing m-aramide (Conex (Trade name) made by Teijin Limited) (relative viscosity IV (in H_2SO_4) = 1.8) in an amount of 10.5 wt%. Next, I cast this dope on a polypropylene film using a coater set to a clearance between the casting blade and film of 30 μm , then caused this to solidify for 5 minutes by a 35°C water/NMP (45/55 weight ratio) solidification bath. I washed this solid by 40°C water, then attached it to a 60°C iron roller and dried it under a constant length to obtain porous film (hereinafter this porous film being referred to as the "Sample 1").

This Sample 1 had a thickness of 13 μm , a gas permeability of 0.9 ml/sec, a rate of retention of gas permeability after heat treatment when treated at 350°C for 10 minutes of 80% of that before treatment, a porosity of 61%, and a specific Young's modulus in the casting direction of 229 $(\text{kgf/mm}^2)/(\text{g/cm}^3)$ and in the direction perpendicular to that of 220 $(\text{kgf/mm}^2)/(\text{g/cm}^3)$. Note that the gas permeability, rate of retention of gas permeability, porosity, and specific Young's modulus were measured in accordance with the methods described in the specification of the present application, pages 32 and 33.

Experiment Regarding the Art of Daido

Aramid Porous Reinforcing Material of Daido

I prepared an aramid porous reinforcing material according to Example 1 of Daido.

Specifically, I added noncrystalline m-aramid continuous filaments of a denier of 3 de as a binder to crystallized m-aramid short filaments of a denier of 1.25 de, used the dry production method to form a film by a basis weight of 19 g/m^2 , and ran this through a calendar roll to obtain a nonwoven fabric sheet (hereinafter this reinforcing material referred to as the "Sample 2").

This Sample 2 had a thickness of 36 μm , a gas permeability of 0.08 ml/sec ($=0.04 \text{ sec}/100 \text{ cc}\cdot\text{in}^2$), a rate of retention of gas permeability after heat treatment when treated at 350°C for 10 minutes of 100% of that before treatment, a porosity of 62%, and a specific Young's modulus in the film-forming direction of 275 (kgf/mm^2)/(g/cm^3). Note that the gas permeability, rate of retention of gas permeability, porosity, and specific Young's modulus were measured in accordance with the methods described in the present specification, pages 32 and 33.

Composite Electrolytic Solution Supporting Polymer Film of Daido

For reference, I prepared a dry composite film relating to the composite electrolytic solution supporting polymer film of Daido. Note that by impregnating this dry composite film with the electrolytic solution as in Example 4, the result becomes the composite electrolytic solution supporting polymer film of Daido.

Specifically, I copolymerized 5.3 mol% of perfluoromethylvinyl ether (PFMV) with polyvinylidene fluoride (PVdF) to obtain a PVdF copolymer. To 15 parts by

weight of this polymer, I added 59.5 parts by weight of dimethyl acetoamide (DMAc) and 25.5 parts by weight of polypropylene glycol having an average molecular weight of 400 (PPG400). I heated and mixed these to dissolve at 60°C to prepare a coating dope. I impregnated and coated the obtained dope in and on said aramid porous reinforcing material (Sample 2) and dipped this film in a DMAc:PPG400:water = 35:15:50 (weight ratio) solidifying solution to solidify the film. Next, I rinsed and dried the film to prepare a dry composite film (hereinafter this dry composite film referred to as the "Sample 3").

SEM Observation

I observed the film cross-sections of the Samples 1 to 3 prepared in the above way by an SEM. Specifically, I prepared observation specimens by freezing by liquid nitrogen and slicing. Further, for the SEM observation, I used an S-2400 made by Hitachi Ltd. to observe the film cross-sections of the Samples 1 to 3.

An SEM photo of the Sample 1 is shown in FIG. 1, an SEM photo of the Sample 2 is shown in FIG. 2, and an SEM photo of the Sample 3 is shown in FIG. 3.

Calculation of Cross-sectional Pore Laminar Coefficient

The cross-sectional pore laminar coefficient was calculated in accordance with the method described in the specification of the present application, page 33. That is, I observed the pores in a region of a length in the longitudinal direction of 5 times the thickness t of the film and designated the maximum length in the sizes of the pores present in the cross-section in the thickness direction as " d ". Next, I calculated the cross-sectional pore laminar coefficient by the relation $(t \cdot p / d)$ based on the thickness t of the film, the porosity p of the porous film, and the maximum pore diameter d .

Specifically, from the SEM photograph of FIG. 1, it is understood that the Sample 1 has a thickness t of the film of 13 μm and a maximum pore diameter d of 0.4 μm , so the Sample 1 was found to have a cross-sectional pore laminar coefficient of 20.

On the other hand, in FIG. 2, the aramid porous reinforcing material is a structure comprised of fibers, so the maximum pore diameter d is difficult to identify from observation of the cross-section. However, if forcibly defining the distance between top and bottom fibers stacked

in the cross-sectional direction as "d", the Sample 2 has a thickness t of the film of 36 μm , a porosity p of 62%, and a maximum pore diameter d of 15 μm , so the cross-sectional pore laminar coefficient is found to be 1.5

Points of Difference Between the Present Invention and the Art of Daido

From the above experimental findings, the present invention and the art of Daido can be said to differ on the following points.

- From FIG. 1, it is understood that the m-aramid-based porous film according to the present invention (Sample 1) is a porous film in form. On the other hand, from FIG. 2, it is understood that the aramid porous reinforcing material according to Example 1 of Daido (Sample 2) is a nonwoven sheet comprised of a large number of fibers randomly superposed. The two can be said to be completely different in structures of materials.

- The m-aramid polymer porous film according to the present invention (Sample 1) can be used in the state as is as a battery separator (see FIG. 1). On the other hand, the m-aramid porous reinforcing material according to Example 1 of Daido (Sample 2) cannot be used in the state as is as a

battery separator (see FIG. 2). Further, Sample 2 can only be used as a battery separator by laminating a PVdF layer over it to obtain a composite film (Sample 3) (see FIG. 3).

- The gas permeability of the aramid porous reinforcing material according to Example 1 of Daido (Sample 2) is 0.08 ml/sec. This is outside the range of "0.2 to 1000 ml/sec" in the present invention as set forth in claim 1. Further, it is also outside of the range of "0.4 to 200 ml/sec" in the present invention as set forth in claim 3.

- The cross-sectional pore laminar coefficient of the aramid porous reinforcing material according to Example 1 of Daido (Sample 2) is 1.5. This is outside the range of "2.5 or more" in the present invention according to claims 1 and 3.

I, the undersigned declarant, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and; further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18, of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 10th day of July , 2007

Satoshi Nishikawa.

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